

# Collective effects in the 2D lattices of the magnetic nanoparticles.

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The work is devoted to investigation of the collective behavior of the regular rectangular lattices of the  $Ni_3Fe$  nanoparticles caused by the dipole-dipole interaction between them. The samples was prepared by the method of the electron lithography and consist of about  $10^5$  particles of  $\sim 50$  nm size. The magnetization curves were investigated by the Hall magnetometry for the different orientation of the external magnetic field at  $4.2^\circ\text{K}$  and  $77^\circ\text{K}$ . The results points on the collective behavior of the system. The observed peculiarities we connect with the quasi-one-dimensional behavior of the system and the formation of the solitons in the system.

The physical properties of arrays of small ferromagnetic particles continue to be an active area of fundamental and theoretical research. The intensive study of those systems was stimulated, in part, by various applications [1]. One of the most interesting effect is the supermagnetic ordering of the system of the interacting ultrafine magnetic particles [2]. In the case of the dipole-dipole interaction, the critical temperature  $T_c$  of the transition in the superparamagnetic state is

$$T_c \sim M^2/R^3,$$

where  $M$  is the magnetization of the particles,  $R$  is the interparticle distance. For example, if the radius of the transition-metal particle is 10nm,  $R \sim 100\text{nm}$ , we have for  $T_c \sim 100^\circ\text{K}$ . Let's mark also, that the type of the long range order in the 2D lattice of the dipole-dipole interacted particles strongly dependences on the symmetry of the lattice due to the long range and anisotropic

character of this interaction. It is known [3,4], that the ground state of the classical 3D dipoles on the rectangular lattice is of the antiferromagnetic type, on the square one is microvortices. The phase of the dipole glass can be the ground state for the random array

For the experimental observation of the supermagnetic transition we must have a regular lattice of the single domain ferromagnetic particles. Moreover the switching time [6] for this particles at  $T \sim T_c$  are found to be less than the time it takes for the measurements. As it is known, the switching time exponentially decreases as the particle volume and coercitive field decreases.

Advances in the nanotechnology make possible the experimental observation of the collective behavior of the small ultrafine ferromagnetic particles. In [7] the experimental observation of the supermagnetic transition in the linear self-assembling mesoscopic *Fe* particle arrays is presented. In [5] the dipolar supermagnetism in the monolayer nanostripes of *Fe* on the vicinal *W*(110) surfaces is reported. By contrast with [7,5] in this letter we carried out the regular 2D rectangular lattices of the nanometer-scale magnetic particles created by the electron lithography method. We observed the unusual magnetic hysteresis for magnetization in the perpendicular direction and investigated main properties of this collective phenomena. It is possible that this effect is determined by the temperature induced nonuniform states which are typical for quasi-one-dimensional systems.

The rectangular lattices of the magnetic nanoparticles are prepared by the electron lithography method from the *Ni<sub>3</sub>Fe* films. The double-layer mask containing the *C<sub>60</sub>* film as a negative electron resist and the *Ti* film as a transmitting layer was used. *Ni<sub>3</sub>Fe* and *Ti* layers are prepared by the laser deposition method at the room temperature. The *C<sub>60</sub>* films were prepared by the sublimation of a the *C<sub>60</sub>* - powder at the temperature 350°C in the vertical reactor with hot walls. The thickness of the masking layers was 20 and 30 nm, accordingly. The thickness of the magnetic layers is varied from 25 nm (samples 2,3) to 45 nm (sample 1). The *C<sub>60</sub>* films is exposed in the JEM - 2000 EXII electron microscope. The parameters of the exposition were the following: the accelerating voltage 200 kV, the exposure dose  $0.05 \div 0.1 C/cm^2$ . The diameter of the electron beam, according to our

estimations, was 300-700 Å. The electron beam irradiation of the  $C_{60}$  films reduces the solubility of the fullerenes in the organic solvents. The most likely reasons of the changes of the solubility are the electron induced polymerization of the  $C_{60}$  molecules accompanied partially graphitization ones [8]. The exposed films were developed in the toluene during 1 min, the  $Ti$  etching was carried out by a plasma etching method in the  $CF_2Cl_2$  atmosphere, the  $Ni$  etching by the ion milling at the  $Ar$  atmosphere. The SEM-images of the one of the samples are shown on the Fig. 1.

The height of the particles is determined by the thickness of the  $Ni_3Fe$  layer. Their diameter dependences, in the main, on the exposed time. The parameters of the investigated samples are summarized in the Table below. There  $a$  and  $b$  are the lattice parameters,  $h$  is the high of the particles and  $d$  is their diameter. The total number of the particles is equal to  $10^5$ .

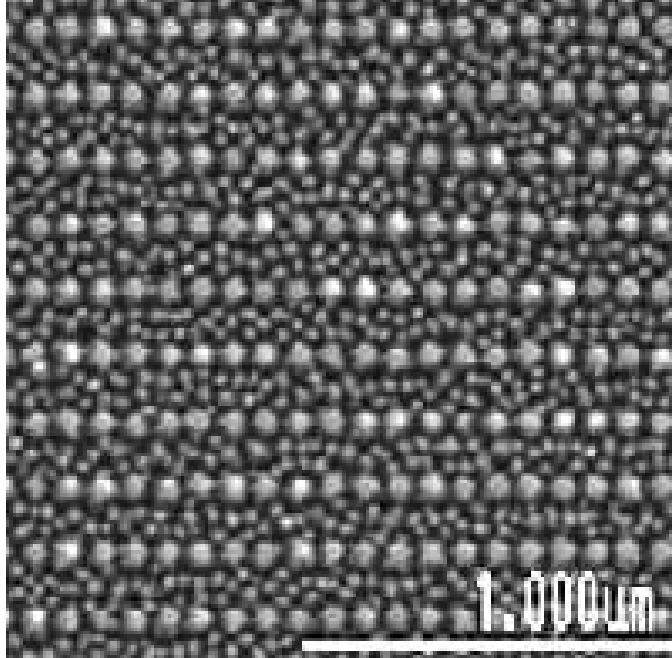


FIG. 1. The SEM-image of the sample 1 (See the table 1). The lattice of the 40-50 nm particles is visible with the background of the 10 nm roughnesses of the sublayer.

N	a(nm)	b(nm)	h(nm)	d(nm)
1	90	180	45	50
2	120	240	25	80
3	120	120	25	70

The measurements of the magnetic properties were provide using the commercial magnetometer based on the Hall response in a semiconductor (InSb). The widths of the current and the voltage probes are 0.2mm and 0.06mm and the thickness of the semiconductor layer is 10 $\mu$ m. The lattice of the particles is located in the active area of one of the Hall crosses. The difference in the Hall voltage between this sample cross and the closely spaced reference is measured using the bridge circuit [9]. Withe the bridge properly balanced, the resulting output voltage is proportional to the sample contribution to the magnetic induction. This contribution can then be calculated so that  $\Delta B = V/RI$  where R is the Hall coefficient and I is the measured current. Typically we use the dc current of 50 mA. The large Hall response in the combination with the good coupling of the small samples to the device results in the excellent spin sensitivity (ratio signal/noise is approximately 100). The sensor works over a large range of the magnetic field and temperature. We investigate the magnetic properties of the samples by measuring the perpendicular magnetization as a function of the direction and magnitude of the applied field. The particles have the polycrystal structure (this was determined by the X-ray and Selected area diffraction) and do not have the anisotropy of the form in the plane of the system. In this case the difference of the magnetization curves for the different orientation of the external magnetic field to the structure lattice is a positive attribute of the collective behavior of the system. As the used method allows to measure only the z-component of the magnetization, we provide our investigation with the thee orientation of the external magnetic field: 1) the field is perpendicular to the sample plane ( $\theta = 0^\circ$ ); 2) the field is

directed at 45 deg. to the sample plane along the short side of the rectangle cell ( $\theta = 45^\circ, \phi = 0^\circ$ );

3) the field is directed at 45 deg. to the sample plane along the long side of the rectangle cell ( $\theta = 45^\circ, \phi = 90^\circ$ ). The results of this measurements for  $T = 4.2K$  are represented on the Figs. 2, 3, 4 accordingly. The difference in the magnetization curves indicates the collective behavior of the system, which is the result of the dipole-dipole interaction between particles. The hysteresis if the field directed at  $\theta = 45^\circ, \phi = 0^\circ$  (Fig. 3) is the attribute of the easy axis of the magnetization which is directed along the short side of the rectangle cell. The remanent magnetization is absent in this case. The existence of such anisotropy in the dipole system was theoretically predicted [4]. The magnetization curves if the field directed at  $\theta = 0^\circ$  or  $\theta = 45^\circ, \phi = 90^\circ$  (Figs. 2, 4) are qualitatively similar. They have hysteresis in the weak magnetic field with the remanent magnetization which is approximately 5 % of the saturation magnetization. The saturation magnetization of the first sample is 35G.

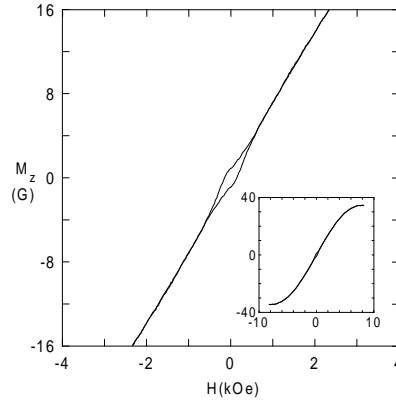


FIG. 2. The dependence of  $M_z$  on the magnetic field with  $\theta = 0^\circ$ . The whole magnetization curve is shown on the casing-in.

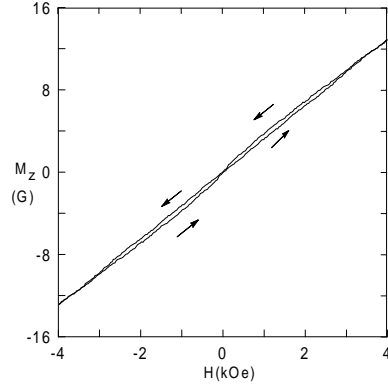


FIG. 3. The dependence of  $M_z$  on the magnetic field with  $\theta = 45^\circ$ ,  $\phi = 0^\circ$ .

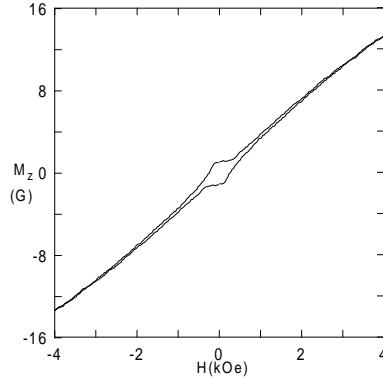


FIG. 4. The dependence of  $M_z$  on the magnetic field with  $\theta = 45^\circ$ ,  $\phi = 90^\circ$ .

The magnetization curves for the second sample is qualitatively similar the those of the first sample, although the samples themselves has the difference shape of the particles. The saturation magnetization of the second one is 18G.

The measurements of the magnetization curves for the system with the square lattice (sample 3), shows the absence of the anisotropy while magnetizing by field directed at  $\theta = 45^\circ$ ,  $\phi = 0^\circ$  or at  $\theta = 45^\circ$ ,  $\phi = 90^\circ$ . This fact approves the isotropy of the single particle in the plane of the system. The hysteresis is absent for any direction of the external field, the remanent magnetization is equal to 0 also. The saturation magnetization of the third sample is 20G.

All results: the existence of the anisotropy axis in the plane of the sample with the rectangular lattice and its absence for the sample with the square lattice were predicted earlier [3] and were principally expected. As for the hysteresis of the magnetization curves and the remanent magnetization for the sample with the rectangular lattice if the external magnetic field direction is  $\theta = 0^\circ$  or  $\theta = 45^\circ$ ,  $\phi = 90^\circ$ , their existence were unexpected. The effect can not be explained by the single particle properties. In this case the remanent magnetization must exist for the every direction of the magnetic field. Besides it must be observed for the sample with the square lattice in this case. Evidently, the existence of the remanent magnetization is the exhibition of the collective properties of the dipole system with the rectangular lattice.

The first sample was investigated at  $T=77K$  also. The hysteresis with the field directed at  $\theta = 45^\circ$ ,  $\phi = 0^\circ$  did not be observed. The hysteresis with the field directed at  $\theta = 0^\circ$  or  $\theta = 45^\circ$ ,  $\phi = 90^\circ$  was qualitatively changed (Fig. 5). This fact directly indicates the dependence of the collective properties on the temperature.

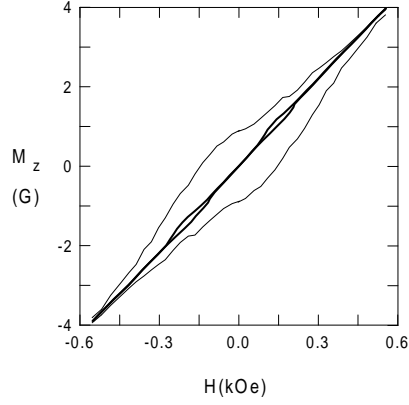


FIG. 5. The changing of the magnetization curve hysteresis with the temperature: the thin line for  $4.2^\circ K$ , the thick one for  $77^\circ K$ .  $H$  is directed at  $\theta = 0^\circ$

Our samples can be considered as a lattices of the interacting single-domain ferromagnetic particles. Model of the 3D classical dipoles is a good approximation for those systems. Energy of the system has the form

$$E = \frac{1}{2} \sum_{\mathbf{r} \neq \mathbf{r}'} D_{ik}(\mathbf{r} - \mathbf{r}') M_i(\mathbf{r}) M_k(\mathbf{r}') + \frac{K}{2} \sum_{\mathbf{r}} M_z^2(\mathbf{r}) - H_i \sum_{\mathbf{r}} M_i(\mathbf{r}),$$

$$D_{ik} = \frac{\delta_{ik}}{r^3} - \frac{3r_i r_k}{r^5}.$$

The numerical simulation were provide for such rectangular (1:2) and square lattices of the dipoles with sizes  $20 \times 50$  and  $30 \times 30$  particles. The system of relaxation Landau-Lifshitz equations were solved with the effective field on the particle equal to the sum of the dipole field of the other particles. The equations were solved with the different values of the single particle anisotropy (the anisotropy axis is perpendicular to the lattice, as our particles are isotropic in the plane of the



system). The relaxation scheme was as follows: at first the randomly directed dipoles relaxed in the external magnetic field which was approximately 1/3 of the saturation field. After that the field was switched off and the following relaxation without external field was performed. The directions of the initial field to the system lattices were similar to those in the experiment. The following results were obtained.

In the case of the square lattice the system relaxes to the microvortex state [3] with  $M_z = 0$  independently on the initial external field orientation.

As for the rectangular lattices, if the initial field was oriented along  $\theta = 45^\circ$ ,  $\phi = 0^\circ$  direction, the final state had the uniform magnetization along the short side of the rectangular. If the initial field has  $\theta = 0^\circ$  or  $\theta = 45^\circ$ ,  $\phi = 90^\circ$  orientation, the system relaxes to the state with the solitons in the chains of the dipoles. They have the antiferromagnetic core with the dipoles oriented perpendicular to chains and ferromagnetic tails in which dipoles oriented along the chains in the opposite directions. The antiferromagnetic core is narrow (it takes only 3-7 cells in the zero magnetic field), but it can have the magnetic moment which value and direction depends on the value of the single particle anisotropy. If the single particle anisotropy is absent, the soliton lies in the plane of the lattice and does not have magnetic moment. With the increase of the single particle anisotropy the soliton obtains the magnetic moment directed along z-axis. For example, the z-component of the soliton magnetic moment is approximately 0.8 of the single particle magnetic moment, if  $K=3$ .

If the anisotropy value is greater than a certain critical one, the system relaxes in the state uniformly magnetized along z-direction independently on the lattice symmetry and initial direction of the external field.

It is necessary the length of the dipoles chains to be at least of approximately 30 dipoles for the stability of the soliton. If the size of the system is less, the soliton, even appearing in the initial field, at zero field ran out from the chain. Evidently, in this case the force attracting the soliton to

the boundary is greater than the pinning force on the discrete lattice. The magnetization reverse of the soliton takes place in the external field much less than the saturation field.

Let us notice especially, that in the case then magnetic moment of the soliton is directed along z-axis, e.i. single particle anisotropy is z-"easy axis", the whole system has the anisotropy of the "easy axis" lying in the plane of the system along the chains of the dipoles (along x-axis), and in the main state dipoles in the chains directed along the chain, but the chains themselves antiferromagnetically ordered.

Using the results of the numerical simulation we suggested the following mechanism of the appearing of the remanent magnetization of our samples with the rectangular lattice. Due to quasi 1D character of the system in the fields near to the saturation the soliton energy is small and their thermoinduced formation is possible. With the decrease of the external field the their energy increases, the width narrows. It leads to their pinning on the discrete lattice. So the remanent magnetization is caused by the existence of the "frozen" solitons, which have formed in the large field. The necessary condition of the z-component of the remanent magnetization of the soliton is the single particle anisotropy. As the particles of the second sample have the disk form, there must be another mechanism of the appearance of the "easy axis" single particle anisotropy (surface, magnetostriction, etc.). The additional experiments are to be carried out to check its existence.

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